

A COMPARISON OF LOW-NO_x BURNERS FOR COMBUSTION OF METHANE AND HYDROGEN MIXTURES

GEIR J. RØRTVEIT,¹ KLAUS ZEPTER,¹ ØYVIND SKREIBERG,¹ MORTEN FOSSUM²
AND JOHAN E. HUSTAD¹

¹*Department of Thermal Energy and Hydropower
The Norwegian University of Science and Technology (NTNU)*

*Kolbjørn Hejes v. 1A
N-7491 Trondheim, Norway*

²*Sintef Energy Research
Sem Sælands v. 11
N-7465 Trondheim, Norway*

Lean premixed fuel mixtures of CH₄ and H₂ are studied in four different low-NO_x burners. These four burners include a fiber burner, a swirl burner, and two porous inert material burners with and without catalytic support. Measurements were performed of the concentrations of NO_x, CO, CO₂, and O₂ in the flue gases. The temperature profiles were also measured for the two different porous burners. The maximum measured temperature was 1830 K. The thermal loads based on the lower heating values were varied from 2.6 to 21 kW, and excess air ratios were varied from 1.0 to 1.9. The NO_x emissions were less than 25 ppm at 3% O₂ for all experiments besides the fiber burner, which under the conditions used had NO_x emissions up to 85 ppm. Another scope of the paper is to clarify the influences of hydrogen addition to methane and its effect on pollutant emissions. For the porous burners hydrogen was found to lower the NO_x emissions slightly, while for the other burners an increase, or no obvious effects, was found. The pressure variations over the burners were also measured, and the swirl burner was found to have the highest pressure drop. The use of low-NO_x burners in gas turbines and industrial boilers is important in the reduction of pollutants from combustion applications. Other applications are small-scale combined heat and power systems utilizing such burners with low-temperature fuel cells or in Stirling engines.

Introduction

The present work explores the influences of different burner concepts on the NO_x and CO emissions in combustion of methane and methane/hydrogen mixtures. The four burners studied include a porous inert material (PIM) burner, a catalytically supported PIM (CSPIM) burner, a radiant surface (fiber) burner from Acotech, and a swirl burner from the International Flame Research Foundation (IFRF). The two latter are commercial burners, while the PIM burner concepts were developed at NTNU, Department of Thermal Energy and Hydropower. The aim of this work is to compare the different burner technologies with respect to emissions of NO_x and CO in addition to differences in pressure drop. A further motivation of this work is accurate testing of the influences of hydrogen addition to methane in the different burners. All experiments presented for 21 kW were performed with natural gas (NG) containing 83% CH₄, 14% ethane, N₂, and CO₂. The term hythane in this paper is used interchangeably for H₂ added to CH₄ and to NG, and “20% H₂” means that 20 vol % of the fuel was substituted with H₂. An emission dependency on excess air ratio (λ) is followed. However, the NO_x

dependencies on maximum measured temperatures are presented and discussed for the PIM burners. Mainly due to the experimental procedures, the numbers for NO_x are given on wet basis while CO values are given on dry basis, both corrected to 3% O₂ in dry flue gases.

Experiments

All burners except the CSPIM were run at thermal loads of 12 and 21 kW in a vertical combustion chamber with 200 mm inner diameter and 600 mm length. The specific heat release rates based on the combustion chamber were thus 0.64 and 1.1 MW/m³, respectively. For the CSPIM the loads were 2.6, 3.3, and 4 kW, giving specific heat release rates of maximum 8.7 MW/m³. The burners and the combustion chambers are cooled by radiation and free convection only.

The Swirl Burner from IFRF

The turbulent diffusion burner in Fig. 1 is an IFRF design. In addition to providing swirl to the

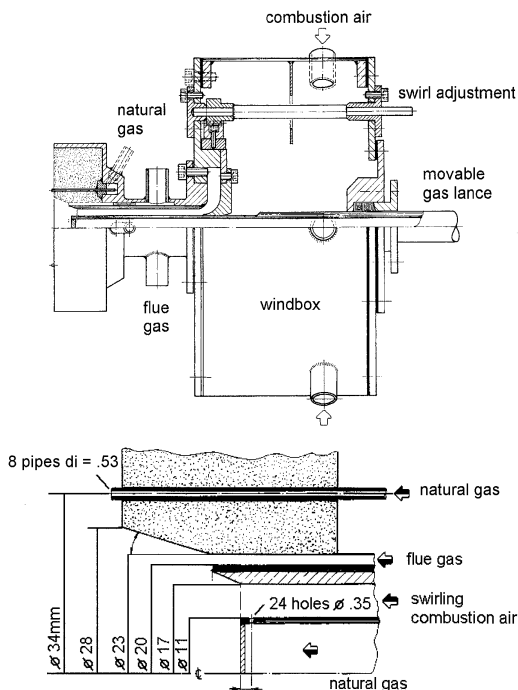


FIG. 1. Schematic illustrations of the swirl burner from IFRF. The bottom illustration gives details of the left-side flame location.

combustion air stream, it gives options to apply external exhaust-gas recirculation and staged combustion by secondary fuel supply. The primary fuel supply position and the swirl number [1] may also be varied. The burner is thus very flexible in terms of operating conditions and can by varying the swirl number operate at low excess air ratios with low emissions due to internal recirculation. The burner loads were kept at 12 and 21 kW.

The Fiber Burner

The fiber burner manufactured by Acotech is presented in Fig. 2. It has a cylindrical shape with an

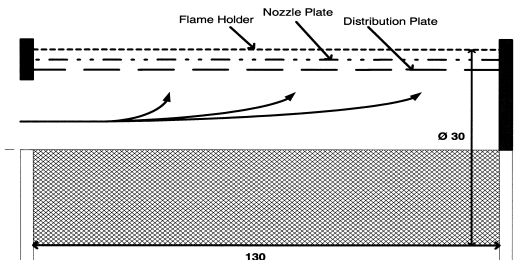


FIG. 2. Schematic illustration of the fiber burner from Acotech.

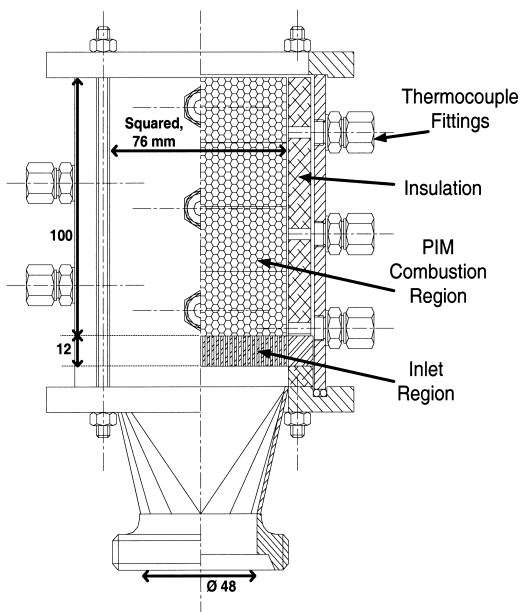


FIG. 3. Schematic illustration of the PIM burner from NTNU.

outer diameter of 30 mm and a length of 130 mm and is designed for premixed operation. The specific heat release rate at 12 kW is 979 kW/m^2 which is above transition to blue-flame combustion mode, where the reaction zone is lifted from the surface and the heat removal by radiation is only 5%–10% of input load [2]. The radiant efficiencies for lower combustion intensities can be as high as 50% according to the manufacturer, giving lower temperatures and thus lower NO_x emissions.

The Porous Inert Media Burner

The PIM burner as seen in Fig. 3 is designed for premixed operation at high excess air ratios. The composite material in the squared 100 mm long main part of the burner is corrugated ceramic fibers ($\text{Al}_2\text{O}_3/\text{SiO}_2$) that provide high heat transport. This high heat transport reduces the temperatures and thus the NO_x emissions and also the specific heat release rates increase by the increased flame speed. High turndown ratio and capability of burning mixtures of low calorific value are additional advantages. The 12 mm long inlet section consists of a perforated ceramic tile with smaller characteristic cavity sizes. This serves as a flame arrestor that extends the stability range by preventing flashbacks. The specific heat release at 12 kW based on the burner dimension is 21 MW/m^3 .

The Catalytically Supported PIM Burner

The Sued Chemie's commercial DO-12 catalyst was tested as the inlet region for a silicon carbide

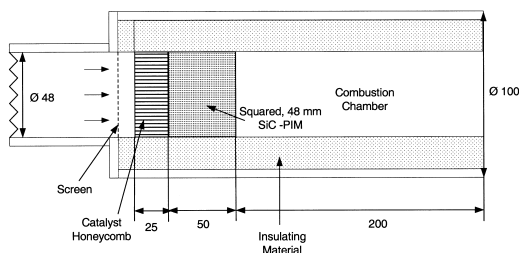


FIG. 4. Schematic illustration of the catalytically supported PIM burner from NTNU.

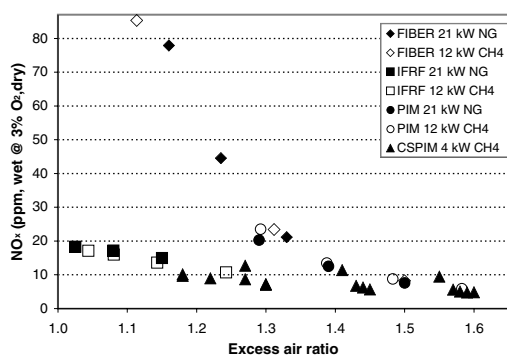


FIG. 5. The NO_x emissions at different excess air ratios for all burners operated with methane or natural gas.

(SiC) PIM burner (refer to Fig. 4). The DO-12 has alumina-based washcoat with Pt and Pd catalysts and is a ceramic honeycomb monolith with 400 cells per square inch. The Reynolds numbers based on the hydraulic diameter in the channels confirm laminar flow. The catalyst was found to completely oxidize hydrogen premixed in air in the range of 1 to 8 vol% H₂. Since oxidation of CH₄ requires an initial temperature for the catalyst of about 700 K, the approach used for igniting was startup by H₂ oxidation. When CH₄ was being oxidized, the amount of hydrogen was lowered. The initial temperature requirement was similar to what is found in ignition curves [3]. Complete combustion of CH₄ was in addition to O₂ and CO₂ measurements verified through total unburned hydrocarbons logging. The specific heat release based on the burner dimension is 23 MW/m³ at 4 kW. The burner stabilized at an operation point, but showed low flexibility toward variation of lambda when gas flow velocities were changed. A successful approach for lowering excess air ratio by substituting air with similar amounts of N₂ is consistently used.

Measurements and Errors

Temperature measurements were made at fixed locations in the PIM and CSPIM. The axial spacing

was 10 mm for the PIM and 20 mm for the CSPIM. The thermocouples were 0.2 mm S-type with a coating to prevent catalytic surface reactions [4]. Due to enclosing of the thermocouples in the matrix of the burner, radiative loss is negligible in the temperature measurements. Maximum temperatures are expected to be within 30 K of the measured maximum temperatures for the PIM and within 80 K for the CSPIM due to the fewer measurement points.

The gas sampling for all experiments was done by two water-cooled stainless-steel measurement probes at the centerline in a combustion chamber placed immediately following the burners. The sampling point for the IFRF, fiber, and PIM was after 450 mm of the total 600 mm length of the combustion chamber. For the CSPIM, the gas sampling was done 20 mm before the exit of its combustion chamber. Preliminary testing showed that the variations over the cross-sections at these two positions were low. The response time for all gas sampling was in order of seconds. NO_x was measured with a chemiluminescent analyzer (Signal NO_x, Series 4000) on wet basis with a heated sampling line, while the sampling line for CO (H&B Uras 3G), O₂ (Sybron, Series 500), and CO₂ (H&B Uras 10P) included a cooler and drierite (CaSi) to remove the water. The calibration gases used for NO_x measurements were 87.4 and 29.8 ppm NO mixtures in N₂. The calibration gases for the CO measurements were 355 and 86 ppm CO mixtures in CO₂ and N₂.

Results and Discussions

Emissions from Methane and Natural Gas Combustion

The NO_x emissions in Fig. 5 show values less than 25 ppm NO_x for all burners, besides the fiber burner. For this burner, the NO_x emissions range up to 85 ppm, but were reduced to 8 ppm when the excess air ratio was increased to 1.5. The numbers for the IFRF decrease from 18 to 11 ppm NO_x as lambda increases from 1.03 to 1.24. The PIM has values ranging from 24 to 6 ppm NO_x for excess air ratios of 1.29 to 1.58. The CSPIM operates with 12 ppm or less NO_x at lambda varying from about 1.2 to 1.6 depending on N₂ dilution amount. All the values presented in Fig. 5 are in the lower range of the NO_x emissions at similar lambda presented in [5] for PIM burners up to 25 kW with different degrees of catalytic support. The EINO_x [6] values vary from 0.2 to 0.9 (g NO₂/kg fuel) for all experiments, except for the fiber burner with values up to 3.0.

The CO emissions for all experiments are given in Fig. 6. What is evident is the low CO levels of 18 ppm or less in most cases. The exception is close to stoichiometric air amounts where the CO increases rapidly for the IFRF burner. The shorter residence time for the CSPIM is most likely the reason

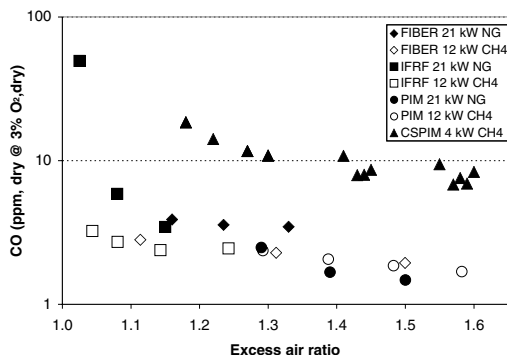


FIG. 6. The CO emissions at different excess air ratios for all burners operated with methane or natural gas.

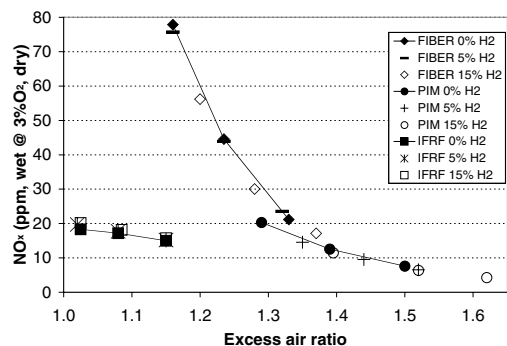


FIG. 7. The NO_x emissions at different excess air ratios for hythane in burners operated at 21 kW.

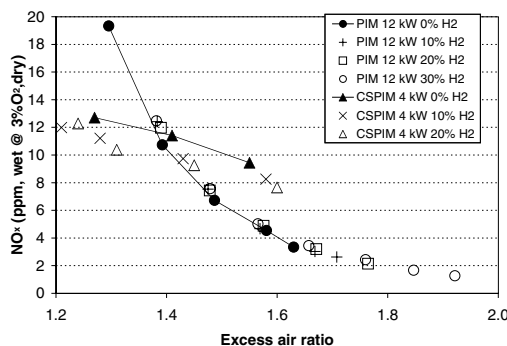


FIG. 8. The NO_x dependencies on excess air ratios from both PIM burners for different hythane fuels.

for these higher CO numbers, and increasing the residence time by lowering thermal load reduces CO. The results show no obvious influence for the emissions from NG versus CH_4 combustion.

The measured maximum temperatures at the fixed points in the PIM section of the CSPIM increased as λ was lowered by increased N_2 dilution up

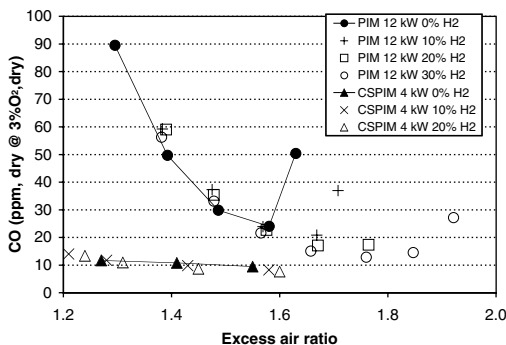


FIG. 9. The CO dependencies on excess air ratio from both PIM burners for different hythane fuels.

to 25 vol% of total flow, although the adiabatic temperatures were kept fairly constant. A temperature profile for normal operation in a PIM burner gives a maximum close to the inlet of the burner section and a decrease due to heat removal toward the exit [7,8]. The higher measured temperatures with increased N_2 dilution were found because the dilution will displace the flame front downstream, while the measurement error was largest close to the inlet region. Adding smaller amounts of CO_2 gave similar results. Dilution with gases like N_2 and CO_2 is of interest due to analogy to exhaust-gas recirculation concepts. A permanent loss of activity was found when the catalyst surface exceeded temperatures of about 1300 K, thus developing a burner application with the use of this catalyst must prevent this. The measured temperatures immediately following the catalyst are less than 900 K for the presented experiments, which ensures long-time durability in stable operation.

Emissions from Hythane Combustion

Figure 7 shows a slight increase of maximum NO_x values for the IFRF burner when substituting some CH_4 with H_2 while keeping a constant thermal load and a constant λ . No clear trends are found for NO_x emissions with H_2 addition at given excess air ratios for the fiber and the PIM at both loads, while the CSPIM shows NO_x values reduced by up to 20% for a constant λ , as seen in Figs. 7 and 8.

The decreasing amounts of carbon in the fuel when adding H_2 could lead to expectations of lower CO emissions. However, the results at 21 kW for CO show no significant effect of adding H_2 to NG. The CSPIM and PIM show no reduction in CO emissions for adding H_2 to the CH_4 at constant λ . The CO numbers for the PIM only are in Fig. 9 measured immediately following the burner without the combustion chamber; these values indicate that the low-emission operating window is enlarged.

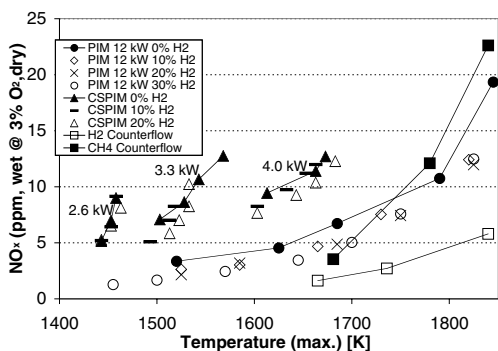


FIG. 10. The NO_x dependencies on temperature for both PIM burners for different hythane fuels.

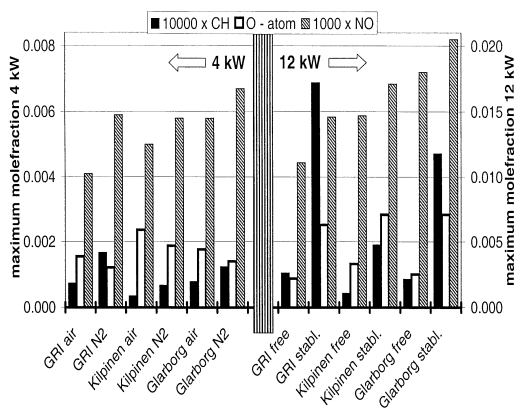


FIG. 11. The maximum radical concentrations for numerical calculations with three different mechanisms. The numbers for 4 kW are comparing a free flame to a N₂-diluted free flame, both with maximum temperatures of 1740 K. The values for 12 kW are comparing a free flame to a stabilized flame, both with $\lambda = 1.4$ and maximum temperature of 1850 K.

The NO_x Formation in the Burners

Moderate temperatures (maximum 1800 K) and thus NO_x formation mainly from the prompt mechanism are verified in the literature [9,10] for swirl burners. According to Ref. [11], the prompt NO_x mechanism is also dominant for the fiber burner at low firing rate and high excess air ratio, while for increasing burner loads and thus temperatures the thermal NO_x becomes significant. Fig. 5 implies the rapid thermal NO_x increase for the fiber burner as λ is lowered and temperatures increased.

The NO_x dependencies on temperature for both PIM burners reveal a fairly constant decrease for each increased amount of H₂ to the CH₄ as in Fig. 10. This constant decrease is consistently found for temperatures below 1800 K where the thermal-NO_x

contribution is insignificant [12], while the reduced carbon contents do not decrease NO_x from the N₂O or the NNH mechanisms. This indicates that the decrease in NO_x is caused by a decrease in the prompt NO_x. According to Ref. [13], a simultaneous increase in radicals like H, OH, and CH₂ will accelerate CH (prompt NO_x) formation, while O₂, H₂O, and CO₂ inhibit its formation. An increase of total NO_x for H₂ addition to CH₄ at constant temperatures was found in a counterflow flame [6]. The increase was because of more H, O, and OH radicals causing more CH in the thin reaction zone. The difference between the curves for numerical calculations of pure H₂ and pure CH₄ counterflow flames [6] given in Fig. 10 is an estimate for the prompt NO_x contribution, while due to no carbon present in H₂ counterflow flames, this curve will include purely NO_x from the thermal, N₂O, or NNH mechanisms. In a porous burner, the reaction zone is wider [8] and the added H₂ seems not to cause more H and OH radicals at the peak temperature where the main part of the CH₄ oxidation occurs. Due to a lower ignition energy, the H₂ oxidation may start at lower temperatures than CH₄ which will widen the reaction zone. The maximum temperatures measured in the experiments for the CSPIM in Fig. 10 are increased with increasing thermal load, which is in agreement with Ref. [8]. Similar trends in NO_x for N₂ dilution at three different temperature levels are another verification of the thermal NO_x insignificance. The small irregularities of the NO_x curves in Fig. 10 occur because the absolute maximum temperatures in the PIM burners have not been found for all cases at the fixed measurement points used, although the irregularities are still within the temperature measurement error. The reason why more NO_x at any given temperature is found in the CSPIM versus the regular PIM may be due to the difference of thermal load.

For the N₂ dilution in the CSPIM, the effect on prompt NO_x is opposite of what is found for the H₂ addition. More N₂ and thus less O₂ leads to increased CH concentrations [13], and also higher concentrations of both CH and N₂ lead to more prompt NO_x through HCN and N in the prompt initiator step. This is consistent with what was found in the experiments in Fig. 10. Computations with CHEMKIN/PREMIX [14] on a free flame analog to the 4 kW experiments for the CSPIM, confirm by substitution of some air with pure N₂ both an increase of CH as seen in the left half of Fig. 11 and also a delay of the flame front. This was found consistently for three different detailed kinetic mechanisms: GRI [15], Kilpinen [16], and Glarborg [17]. The same calculations verify that NO_x from the thermal, N₂O, and NNH mechanisms contribute to less than about 25% of total NO_x both with and without N₂ dilution at these temperatures.

Characteristic for combustion in a PIM burner is the higher flame speed due to enhanced heat transport to the incoming gas mixture. Computations

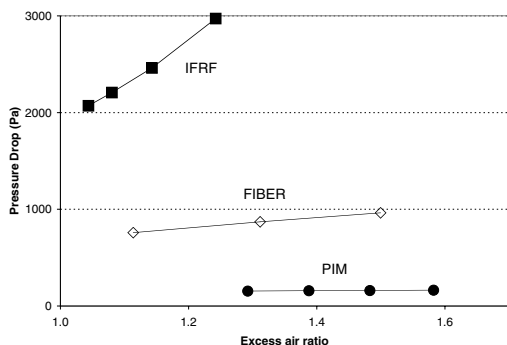


FIG. 12. The pressure-drop dependencies on excess air ratio for the different burners at 12 kW.

were performed by forcing the high experimental gas-flow rates on a fixed free-flame temperature profile. The results with the three different mechanisms reveal consistent large increases of the CH concentrations in stabilized flames compared to free flames indicating more prompt NO_x in PIM burners (refer to the right half of Fig. 11). Fig. 11 also presents the more moderate increases of both O and NO concentrations.

Thus both from the previous discussion and from Figs. 10 and 11, it is concluded that prompt NO_x is by far most significant in PIM burners, which is consistent with what is found in Ref. [18]. A decrease in both radicals and NO_x with increasing amounts of flue gas recirculation at constant lambda and thermal load is found in Ref. [19]. This apparent difference to the results presented here is due to temperature reduction and possibly the inhibition effect on CH by H_2O [13] occurring both in their burner and in normal recirculation concepts.

Pressure Drop for the Burners

The fan power needed to achieve the appropriate flow conditions and mixing through a burner, and therefore the system efficiency, is given by the pressure drop. The total pressure drops including the mixer upstream of the burners are given for the pre-mixed PIM and fiber burners. A pressure drop of 160 Pa over the PIM at a thermal load of 12 kW, according to Fig. 12, requires a net fan power of less than 1 W. The CSPIM is assumed to have a pressure drop approximately similar to the regular PIM, and the pressure drop range is up to 1000 Pa for the fiber burner. The swirl burner from IFRF with a pressure drop up to 3000 Pa needs a net fan power of 8 to 14 W, depending on lambda.

Conclusions

The NO_x and CO emissions were found to be significantly dependent on burner type and excess air

ratio. Hydrogen addition to natural gas or methane was found beneficial for stability purposes in the PIM burners, but was not found to have a considerable effect on lowering emissions in any of the burners run at 12 and 21 kW when compared at a constant lambda. Hydrogen addition was found beneficial in the CSPIM burner where decreasing NO_x was found for a constant lambda. NO_x emissions lower than 25 ppm at 3% O_2 were found for all burners except for the fiber burner. The significance of prompt NO_x was verified for these low- NO_x burners, and a lowered prompt NO_x contribution for the PIM burners was found for increasing H_2 addition at a constant temperature. The IFRF swirl burner shows promising low emissions of both NO_x and CO at low excess air, but its high pressure drop must be taken into consideration in an application. The fiber burner shows better system efficiencies and also low CO emissions, but the increased NO_x emissions as lambda is lowered indicate that this burner must be operated at lower thermal loads. The PIM burner has a very low pressure drop and low emissions of both NO_x and CO, but must be run at higher excess air ratios. The excess air ratios may be lowered by dilution for the CSPIM burner while keeping the NO_x and CO emissions low.

Acknowledgments

We are indebted to Anders Strømman for his participation in some of the experimental work. This research was supported financially by the Norwegian Research Council.

REFERENCES

- Beer, J. M., and Chigier, N. A., *Combustion Aerodynamics*, Applied Science Publishers, London, 1972, p. 106.
- Kulkarni, M. R., and Peck, R. E., *Joint Mtg. Canadian Western States Sect. Combust. Inst.* 2:98 (1990).
- Deutschmann, O., Maier, L. I., Riedel, U., Stroemman, A. H., and Dibble, R. W., *Catal. Today* 59:141 (2000).
- Kent, J. H., *Combust. Flame* 14:279 (1970).
- Cerri, I., Saracco, G., and Specchia, V., *Catal. Today* 60:21 (2000).
- Rørtveit, G. J., Hustad, J., and Williams, F. A., *Int. Conf. Technol. Combust. Clean Air* 6:339 (2001).
- Zepter, K., and Hustad, J., *Proc. Scan.-Nordic Sect. Combust. Inst.* 1:275 (2001).
- Brenner, G., Pickenacker, K., Pickenacker, O., Trimis, D., Wawrzinek, K., and Weber, T., *Combust. Flame* 123:201 (2000).
- Claypole, T. C., and Syred, N., *Proc. Combust. Inst.* 18:81 (1980).
- Toqan, M. A., Beer, J. M., Jahsohn, P., Sun, N., Testa, A., Shihadeh, A., and Teare, J. D., *Proc. Combust. Inst.* 24:1391 (1992).

11. Røkke, N. A., "Experimental and Theoretical Studies of Environmental Aspects of Natural Gas Combustion," Ph.D. thesis, Norwegian University of Science and Technology, Trondheim, 1994.
12. Turns, S. R., *Prog. Energy Combust. Sci.* 21:361 (1995).
13. Li, S. C., and Williams, F. A., *Combust. Flame* 118:399 (1999).
14. Kee, R. J., Rupley, F. M., Miller, J. A., Coltrin, M. E., Grcar, J. F., Meeks, E., Moffat, H. K., Lutz, A., Dixon-Lewis, G., Smoke, M. D., Warnatz, J., Evans, G. H., Larson, R. S., Mitchel, R. E., Petzold, L. R., Reynolds, W. C., Caracotsios, M., Steward, W. E., and Glarborg, P., *CHEMKIN Collection 3.5*, Reaction Design, San Diego, 1999.
15. Smith, G. P., Golden, D. M., Frenklach, M., Moriarty, N., Eiteneer, B., Goldenberg, M., Bowman, C. T., Hanson, R. K., Song, S., Gardiner Jr., W., Lissianski, V. V., and Qin, Z., GRI-MECH Home Page, Gas Research Institute, 2000, www.me.berkeley.edu/gri_mech/.
16. Coda Zabetta, E., Kilpinen, P., and Hupa, M., *Energy Fuels* 14:751; 14:1335 (2000).
17. Glarborg, P., Alzueta, M. U., Dam-Johansen, K., and Miller, J. A., *Combust. Flame* 115:1 (1998).
18. Pickenacker, O., Trimis, D., and Durst, F., *Eur. Conf. Small Burner Heating Technol.* 1:11 (2000).
19. Mößbauer, S., Gruber, W., and Trimis, D., *Int. Conf. Technol. Combust. Clean Air* 6:709 (2001).